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(54) ORGANIC EL DISPLAY

ORGANISCHE ELEKTROLUMINESZIERENDE ANZEIGEVORRICHTUNG

AFFICHEUR ELECTROLUMINESCENT ORGANIQUE

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(56) References cited:
EP-A- 0 892 589 **DE-A1- 19 501 229**
JP-A- 7 272 857 **JP-A- 10 162 958**
JP-A- 10 289 784 **JP-A- 11 185 955**

- **PATENT ABSTRACTS OF JAPAN vol. 1998, no. 11, 30 September 1998 (1998-09-30) -& JP 10 162958 A (CASIO COMPUT CO LTD), 19 June 1998 (1998-06-19)**
- **KNOLL R.W.; HENAGER JR C.H.: "Optical and physical properties of sputtered Si:Al:O:N films", JOURNAL OF MATERIALS RESEARCH, vol. 7, no. 5, May 1992 (1992-05), pages 1247-1252, XP009113651, MATERIALS RESEARCH SOCIETY, WARRENDALE, PA, USA**

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Description

Technical Field

[0001] The present invention relates to an organic EL display device, particularly an organic EL display device wherein the quantity of EL emission that can be taken out is large.
[0002] The "EL" described in claims and so on in the present description is an abbreviated representation of "electroluminescence".

Background Art

[0003] Hitherto, attention has been paid to the utilization of EL displays using electroluminescence as luminescent elements in various display devices since the EL display devices have characteristics that the EL elements are capable of emitting light for itself, the capability of being watched and perceived is high, and they are superior in impact-resistance because of the perfect solid state thereof. Particularly, organic EL display devices using an organic compound as their luminescent material have been positively made practicable since the devices make it possible to lower voltage to be applied largely and can easily be made thin and small-sized so that the consumption power thereof can be made small.
[0004] Such an organic EL display device 100 is disclosed in JP-Laid/Open-Hei-,10-289784 and JP-Laid/Open-Hei-11-185955. The outline of this device 100 is illustrated in FIG. 5. An organic EL element 130 is composed to sandwich an organic compound containing a luminescent layer 124 between a lower electrode (conductive layer) 122 deposited on a supporting substrate 121 and an upper electrode (hole injection electrode) 125, and a sealing member 126 for excluding the effect of moisture in the atmosphere is set above this organic EL element 130. Furthermore, a color filter layer 127 is disposed on the face, opposite to the organic EL element 130, of the sealing element 126. In the example of this organic EL element 130, a nonconductive layer 123 is arranged between the lower electrode 122 and the luminescent layer 124. A void (for example, gas such as nitrogen) layer 131 is present between the color filter layer 127 and the upper electrode 125.
[0005] Accordingly, by applying a given voltage between the upper and lower electrodes 122 and 125, EL emission passes from the side of the upper electrode 125, which is a transparent electrode, through the void layer 131, the color filter layer 127 and the sealing member 126 and then the EL emission can be taken out. In FIG. 5, an arrow represents the direction in which the EL emission is taken out.
[0006] As illustrated in FIG. 6, JP-Laid/Open-Hei-10-162958 discloses an organic EL display device 200 wherein color changing layers 201 and 202, a protective layer 203, a transparent electrode 204, an organic luminescent layer 205 and a rear electrode 220 are disposed below an insulating substrate (glass substrate) 210. The device 200 is composed so as to take out EL emission through the color changing layers 201 and 202 from the side of the transparent electrode 204.
[0007] Therefore, by applying a given voltage between the upper and the lower electrodes 204 and 220, EL emission passes from the side of the transparent electrode 204 through the protective layer 203, the color changing layers 201 and 202, and the insulating substrate 210 and then the EL emission can be taken out. In FIG. 7, an arrow represents the direction in which the EL emission is taken out.
[0008] In the case that light passes through an interface between two layers a and b made of constituent materials having different refractive indexes, the relationship between the reflectivity R of the interface (the reflectivity against light perpendicular to the interface) and the refractive indexes n_a and n_b of the constituent materials of the two layers is represented by the following:

$$R = (n_a - n_b)^2/(n_a + n_b)^2$$

[0009] As can be understood from this expression, therefore, the reflectivity R of the interface becomes larger as the difference between the refractive indexes n_a and n_b of the constituent materials of the two layers becomes larger. As a result, the quantity of the light transmitting through the interface decreases.
[0010] For example, when light is radiated from an indium zinc oxide (IZO, refractive index: 2.1) and the light comes through a void layer (refractive index: 1.0) into a glass substrate (refractive index: 1.5), the light quantity incoming in the glass substrate is reduced to 84% of the light quantity (100%) outgoing from the IZO on the assumption that the light absorbance of the respective layers themselves are zero.
[0011] However, according to the organic EL display devices disclosed in JP-Laid/Open-Hei-10-289784 and 11-185955, as the transparent conductive material constituting the upper electrode, an indium tin oxide (ITO) having a refractive index of about 2 or the like material is used, and the refractive index of the void (gas such as nitrogen) layer between the upper electrode and the sealing member is 1. Therefore, the refractive index difference between the upper

electrode and the void layer and the refractive index difference between the void layer and the sealing member become large. In general, the color changing layers are also made of a polymer material having a far larger refractive index value than that of the void layer. For this reason, the refractive index difference between the void layer and the color changing layers also becomes large. This results in a problem that EL emission reflects on the respective interfaces so that the quantity of the EL emission which can be taken out becomes remarkably small.

[0012] In the organic EL display device disclosed in JP-Laid/Open-Hei-10-162958, the relationship between the refractive indexes of the respective layers is not considered. Therefore, EL emission reflects on the respective interfaces. The number of the layers through the EL emission must transmit is large. These facts result in a problem that the quantity of the EL emission which can be taken out gets less.

[0013] JP-Laid/Open-Hei-7-272857 discloses an inorganic EL element formed on a supporting substrate and having a structure wherein, on its upper electrode side, a silicone oil (sealing agent) having a refractive index (s2) smaller than the refractive index (s1) of the upper electrode and a protective layer having a refractive index (s3) smaller than that of this silicone oil and larger than 1 are disposed and further EL emission is taken out from the side of the upper electrode.

[0014] However, when the silicone oil used in this inorganic EL element is used as a sealing agent of an organic EL element, the silicone oil may cause an organic luminescent medium in the organic EL element to be dissolved and invades into its layer interface to disturb its layer structure. Thus, the organic EL element may be deteriorated and the durability thereof may also be deteriorated.

[0015] In such an inorganic EL element, the luminescent brightness thereof is originally low. Thus, even if each of the refractive indexes of its upper electrode, sealing agent and protective layer is considered, it is practically difficult that the inorganic EL element exhibits performance equivalent to that of organic EL elements or is easily produced.

[0016] Thus, the inventors of the present invention have found out that by considering the relationship between the refractive indexes of a sealing member and a color changing medium and those of a transparent electrode and so on, the quantity of EL emission which can be taken out in an organic EL display device can be made large.

[0017] That is, an object is to provide an organic EL display device wherein even if a sealing member is set up and EL emission is taken out through the sealing member reflection on each of interfaces is effectively suppressed so that the quantity of the EL emission which can be taken out is large.

Disclosure of the Invention

[0018] According to the present invention, there is provided an organic EL display device comprising; a supporting substrate; an organic EL element comprising an organic luminescent medium sandwiched between a lower electrode and an upper electrode on the supporting substrate; a sealing medium; and a sealing member; EL emission being taken out from the upper electrode; wherein a following expression (1) is satisfied

$$n1 \geqq n2 \geqq n3 \qquad (1)$$

wherein n1 represents a refractive index of the upper electrode, n2 represents a refractive index of the sealing medium, and n3 represents a refractive index of the sealing member.

[0019] It is sufficient for the present invention that the expressions about the refractive indexes described above are satisfied by at least the peak wavelength of EL emission or conversion light.

[0020] That is, such a structure makes it possible that even if the sealing member is provided and EL emission is taken out through the sealing member, reflection on each interface is suppressed so as to provide an organic EL display device having a large luminescence quantity.

[0021] In the organic EL display device, it is preferred that the device further comprises a color filter and/or a fluorescent medium, that is, a color changing medium (which may be referred to as a first color changing medium) between the sealing medium and the sealing member; and a following expression (2) is satisfied

$$n1 \geqq n2 \geqq n4 \geqq n3 \qquad (2)$$

wherein n4 represents a refractive index of the color changing medium.

[0022] Such a structure makes it possible that even if the sealing member is provided and further the first color changing medium is provided to display an image, reflection on each interface is suppressed so as to provide an organic EL display device having a large luminescence quantity.

[0023] In the organic EL display device, it is preferred that the device further comprises a color changing medium (which may be referred to as a second color changing medium) on a surface of the sealing member, the surface opposite to a surface on which the sealing medium is arranged; and a following expression (3) is satisfied

$$n1 \geqq n2 \geqq n3 \geqq n4' \qquad (3)$$

wherein n4' represents a refractive index of the color changing medium.

[0024] Such a structure makes it possible that even if the sealing member is provided and further the second color changing medium is provided to display an image, reflection on each interface is suppressed so as to provide an organic EL display device having a large luminescence quantity. Since the second color changing medium does not contact the sealing medium directly, the second color changing medium may not be deteriorated by the sealing medium. Moreover, it is possible to prevent the generation of snapping or the like resulting from irregularities on the surface of the second color changing medium.

[0025] In the organic EL display device as formed, it is preferred that the refractive indexes n1 and n2 satisfy a following expression (4):

$$n1 \geqq n2 \geqq 0.7 \times n1 \qquad (4)$$

[0026] Such a structure makes it possible to provide an organic EL display device having a larger luminescence quantity since the refractive index n1 of the upper electrode and the refractive index n2 of the sealing medium become nearer to each other.

[0027] In the first organic EL display device, it is preferred that the refractive index of the sealing medium is 1.56 or more.

[0028] Such a structure reduces danger of deterioration of the organic EL display device by the sealing medium and allows the selection of materials of the upper electrode and the sealing member among a wider range of various materials. A typical example of a sealing liquid having a refractive index of less than 1.56 is silicone oil. In the case that such silicone oil is used, this sealing liquid causes deterioration of the organic luminescent medium so that the durability thereof may decrease.

[0029] In the organic EL display device, the sealing medium comprises a transparent resin and/or a sealing liquid.

[0030] Such a structure makes it possible to prevent display defects based on light scattering since the sealing medium can be handled with less influence by taking-in of bubbles and so on.

[0031] In the organic EL display device, it is preferred that the sealing medium comprises a transparent inorganic material.

[0032] Such a structure makes it possible to improve the reliability of the organic EL display device without hindering luminescence of the organic EL and without oxidizing or deteriorating the organic EL element readily since the transparent inorganic material does not comprise water content, oxygen or low molecular monomers and has the higher blocking effect.

[0033] The wording "comprises a transparent inorganic material" includes the meaning of a transparent inorganic film or the material dispersed in the above-mentioned transparent resin or sealing liquid.

[0034] In the organic EL display device, it is preferred that the upper electrode is made mainly of an indium zinc oxide.

[0035] The use of the indium zinc oxide in the upper electrode makes it possible to satisfy the above-mentioned expression (1) or (2) since the refractive index of the indium zinc oxide is as high as about 2.1.

[0036] In the organic EL display device, it is preferred that a thin film transistor (hereinafter referred to as a TFT) for driving the organic EL element is arranged on the supporting substrate.

[0037] Such a structure allows significantly low driving voltage with enhanced luminescence efficiency and lower power consumption.

Brief Description of Drawings

[0038]

FIG. 1 is a graph showing relationship between the refractive index of a sealing medium and the transmittance thereof.
FIG. 2 is a sectional view of an organic EL display device in a first embodiment.
FIG. 3 is a circuit diagram of one example of active driving type organic EL display devices.
FIG. 4 is a perspective view in a plane direction of the active driving type organic EL display device according to the circuit diagram illustrated in FIG. 3.
FIG. 5 is a sectional view of a conventional organic EL display device (No. 1).

FIG. 6 is a sectional view of a conventional organic EL display device (No. 2).

Best Mode for Carrying Out the Invention

[0039] Embodiments of the present invention will be specifically described with reference to the drawings. In the drawings which are referred to, the size of respective constituent components, the shape thereof and the arrangement relationship therebetween are roughly illustrated in such a manner that this invention can be merely understood. Therefore, this invention is not limited to illustrated examples. In the drawings, hatching, which represents sections, may be omitted.

[0040] As illustrated in FIG. 2, the first embodiment is specifically an active matrix type organic EL display device 62 wherein TFTs 14 embedded in an electrically insulator (including a gate insulating film) 12, an inter-insulator (flattening film) 13, organic EL elements 26 and contact holes (electrically connecting members) 48 for connecting the TFTs 14 and the organic EL elements 26 electrically are disposed on a supporting substrate (which may be referred to only as a substrate) 10. The device 62 further comprises a sealing medium 16, a color changing medium 60 and a sealing member 58.

[0041] The refractive index of an upper electrode 20 in the organic EL element 26, that of the sealing medium 16 and that of the sealing member 58 are represented by n1, n2 and n3, respectively. In this case, the following expression (1) is satisfied in the active matrix type organic EL display device 62.

$$n1 \geqq n2 \geqq n3 \qquad (1)$$

[0042] The following will describe the constituent elements and so on in the first embodiment, referring to FIG. 2 and so on.

1. Refractive index

① Expression (1)

[0043] In the organic EL display device, the refractive indexes of the respective layers satisfy at least the expression (1), as described above.

[0044] That is, by satisfying the expression (1) in this way, the reflectance of EL on interfaces between the respective layers can be reduced so that an organic EL display device having a large luminescence quantity can be provided.

[0045] For example, when light is radiated from the upper electrode made of an indium zinc oxide (IZO, refractive index: 2.1) and the light comes through the sealing medium (refractive index: 1.7) into the sealing member (refractive index: 1.5), the quantity of the light which can come in the sealing member becomes a high value, such as 98.6% of the quantity (100%) of the light outgoing from the upper electrode on the assumption that the light absorbance of the respective layers themselves are 0%. In other words, it can be understood that light hardly reflects on the respective interfaces.

[0046] Assuming, as a model, a case in which light is radiated outside from the side of the upper electrode (IZO, refractive index: 2.1) and then the light comes through the sealing medium into the sealing member (refractive index: 1.5) in the organic EL element made illustrated in FIG. 2, the relationship between the refractive index of the sealing medium and the quantity of the light incoming in the sealing member can be shown as in FIG. 1. Namely, in FIG. 1, its transverse axis represents the refractive index value of the sealing medium and its vertical axis represents the transmittance (%) of the sealing medium, that is, the percentage of the quantity of the light incoming in the sealing member. For example, when this transmittance is 100(%), it is meant that all EL emission taken out from the organic EL element comes in the sealing member without being absorbed in the sealing medium or reflected thereon.

[0047] As can be understood from FIG. 1, when the expression (1) is satisfied and the refractive index of the sealing medium becomes nearer to the refractive index value of the upper electrode, the percentage of the quantity of the light incoming in the sealing medium trends to be larger.

[0048] In this example, therefore, a high transmittance of 97% or more can be obtained in the sealing medium by satisfying the expression (1).

[0049] The refractive index is defined as a refractive index relative to 1 as the value of vacuum. If interfaces between the respective layers of the upper electrode, the sealing medium and the sealing member are mixed with each other to be indefinite, the mixed layer is defined as an average refractive index. However, even if the mixed layer is present, it is preferred that the average refractive index becomes smaller successively from the upper electrode to the sealing member.

② Expression (2)

[0050] As illustrated in FIG. 2, when the first color changing medium 60 is arranged between the sealing medium 16 and the sealing member 58 and further the refractive index of the first color changing medium 60 is represented as n4 in the organic EL display device 62 of the first embodiment, it is more preferred to satisfy the above-mentioned expression (1) and further satisfy the following expression (2).

$$n1 \geqq n2 \geqq n4 \geqq n3 \qquad (2)$$

[0051] When in such a structure, light is radiated from the upper electrode made of, for example, an indium zinc oxide (IZO, refractive index: 2.1) and then the light comes through the sealing medium (refractive index: 1.6) and the first color changing medium (refractive index: 1.55) into the sealing member (refractive index: 1.5), the quantity of the light which can come in the sealing member becomes a high value of 98% of the light quantity (100%) outgoing from the upper electrode on the assumption that the light absorbance of the respective layers themselves are 0%.

③ Expression (3)

[0052] When the second changing medium is disposed on the side, opposite to the sealing medium, of the sealing member, that is, the side which contacts the air and further the refractive index of the second color changing medium represented as n4' in the organic EL display device of the first embodiment, it is more preferred to satisfy the above-mentioned expression (1) and further satisfy the following expression (3).

$$n1 \geqq n2 \geqq n3 \geqq n4' \qquad (3)$$

[0053] When in such a structure, light is radiated from the upper electrode made of, for example, an indium zinc oxide (IZO, refractive index: 2.1) and then the light comes through the sealing medium (refractive index: 1.7) and the sealing member (refractive index: 1.55) into the second color changing medium (refractive index: 1.5), the quantity of the light which can come in the second color changing medium becomes a high value of 99% of the light quantity (100%) outgoing from the upper electrode on the assumption that the light absorbance of the respective layers themselves are 0%.

④ Expression (4)

[0054] In the organic EL display device, it is preferred that the refractive index n1 of the upper electrode and the refractive index n2 of the sealing medium satisfy the above-mentioned expression (1) and further satisfy the following expression (4).

$$n2 \geqq 0.7 \times n1 \qquad (4)$$

[0055] When in such a structure, light is radiated from the upper electrode made of, for example, an indium zinc oxide (IZO, refractive index: 2.1) and then the light comes through the sealing medium (refractive index: 1.55) into the sealing member (refractive index: 1.5), the quantity of the light which can come in the sealing medium becomes a high value of 98% of the light quantity (100%) outgoing from the upper electrode on the assumption that the light absorbance of the respective layers themselves are 0%.

2. Substrate

(1) Kind

[0056] The substrate (which may be referred to as the supporting substrate) in the organic EL display device is a member for supporting the organic EL elements, TFTs and so on. It is therefore preferred that the mechanical strength and the dimensional stability thereof are superior.

[0057] Examples of such a substrate are substrates made of an inorganic material, such as a glass plate, a metal plate, and a ceramic plate. Preferred examples of the inorganic material include glass material, silicon oxide, aluminum oxide, titanium oxide, yttrium oxide, germanium oxide, zinc oxide, magnesium oxide, calcium oxide, strontium oxide, barium oxide, lead oxide, sodium oxide, zirconium oxide, sodium oxide, lithium oxide, boron oxide, silicon nitride, soda lime glass, barium/strontium-containing glass, lead glass, aluminosilicate glass, borosilicate glass, and barium borosilicate glass.

[0058] Preferred examples of an organic material constituting the substrate include polycarbonate resin, acrylic resin, vinyl chloride resin, polyethylene terephthalate resin, polyimide resin, polyester resin, epoxy resin, phenol resin, silicone resin, fluorine resin, polyvinyl alcohol resin, polyvinyl pyrrolidone resin, polyurethane resin, epoxy resin, cyanate resin, melamine resin, maleic resin, vinyl acetate resin, polyacetal resin and cellulose resin.

(2) Surface treatment, and so on

[0059] In order to avoid the invasion of moisture into the organic EL display device, it is also preferred that the substrate made of any one of these materials is subjected to moisture-proof treatment or hydrophobic treatment by forming an inorganic film or applying fluorine resin.

[0060] This is particularly effective when an organic material such as a polymer is used.

[0061] In order to avoid the invasion of moisture into the organic luminescent medium, it is preferred to make the water content in the substrate and the gas permeability coefficient thereof small. Specifically, it is preferred that the water content in the supporting substrate is 0.0001% by weight or less and the gas permeability coefficient is 1×10^{-13} cc • cm/cm^2 • sec.cmHg or less.

[0062] In order to take out EL emission from the side of the upper electrode in the first embodiment, it is unnecessary that the substrate itself has transparency. However, in the case that EL emission is taken out through the substrate (also in the case of use as the sealing member), it is preferred to use, among the above-mentioned substrate materials, the substrate material having a light transmittance of 70% or more particularly at a wavelength of 400 to 700 nm.

(3) Refractive index

[0063] It is preferred to set the refractive index of the substrate in the range of 1.4 to 1.8. This is because by setting the refractive index within such a range the constituent material of the substrate which can be used can be selected from wider range of materials.

[0064] This is also because by setting the refractive index of the substrate in such a range, the above-mentioned expressions can be satisfied in the relationships to the refractive index of the upper electrode and that of the lower electrode.

[0065] Furthermore, this is because such a substrate makes it possible to suppress reflection on the surface of the substrate, even if EL emission is taken out through the substrate.

[0066] For reference, the values of the refractive indexes of preferred substrates are as follows:

methyl methacrylate resin : 1.49
silicon oxide (SiO_2) : 1.54
boron oxide (B_2O_3) : 1.77
glass : 1.49-1.50
tetrafluoroethylene resin : 1.49

3. Organic EL element

(1) Organic luminescent medium

[0067] The organic luminescent medium can be defined as a medium containing an organic luminescent layer wherein an electron and a hole are recombined with each other so that EL can be emitted. This organic luminescent medium can be made, for example, by depositing the following layers on an anode:

① organic luminescent layer
② hole injection layer/organic luminescent layer
③ organic luminescent layer/electron injection layer
④ hole injection layer/organic luminescent layer/electron injection layer
⑤ organic semiconductor layer/organic luminescent layer
⑥ organic semiconductor layer/electron barrier layer/organic luminescent layer
⑦ hole injection layer/organic luminescent layer/adhesion improving layer

[0068] Among these, the structure ④ is preferably used since it can give a higher luminescent brightness and is also superior in durability.

① Constituent material

[0069] Examples of the luminescent material in the organic luminescent medium include one or a combination of two or more selected from p-quaterphenyl derivatives, p-quinquphenyl derivatives, benzothiazole compounds, benzoimidazole compounds, benzoxazole compounds, metal-chelated oxinoid compounds, oxadiazole compounds, styrylbenzene compounds, distyrylpyrazine derivatives, butadiene compounds, naphthalimide compounds, perylene derivatives, aldazine derivatives, pyrazine derivatives, cyclopentadiene derivatives, pyrrolopyrrole derivatives, styrylamine derivatives, coumarin compounds, aromatic dimethylidyne compounds, metal complexes having a ligand of a 8-quinolynol derivative, and polyphenyl compounds.
[0070] Among these organic luminescent materials, 4,4'-bis(2,2-di-t-butylphenylvinyl)biphenyl (abbreviated to DTBPBBi) and 4,4'-bis(2,2-diphenylvinyl)biphenyl (abbreviated to DPVBi) as aromatic dimethylidyne compounds, and derivatives thereof are more preferred.
[0071] Furthermore, it is preferred to use a material wherein an organic luminescent material having a distyrylarylene skeleton or the like, as a host material, is doped with a fluorescent dye giving intense blue and red fluorescence, for example, a coumarin material, or a fluorescent dye similar to the host, as a dopant. More specifically, it is preferred to use the above-mentioned DPVBi or the like as a host and use N,N-diphenylaminobenzene (abbreviated to DPAVB) as a dopant.
[0072] It is also preferred to use a high molecular material (number average molecular weight: 10000 or more) as well as the above-mentioned low molecular material (number average molecular weight: less than 10000).
[0073] Specific examples thereof include polyarylene vinylene, derivatives thereof (PPV), polyfluorene, derivatives thereof, and fluorene-containing copolymers.

② Thickness

[0074] The thickness of the organic luminescent medium is not particularly limited. It is however preferred to arrange the thickness in the range of, for example, 5 nm to 5 μm.
[0075] The reasons for this are as follows: if the thickness of the organic luminescent medium is below 5 nm, the luminescent brightness and durability thereof may deteriorate, and if the thickness of the organic luminescent medium is over 5 μm, the value of the voltage to be applied may become high.
[0076] Therefore, the thickness of the organic luminescent medium is more preferably in the range of 10 nm to 3 μm, and is still more preferably in the range of 20 nm to 1 μm.

(2) Electrodes

[0077] The following will describe an anode layer and a cathode layer as the electrodes. The anode layer and the cathode layers become the upper and the lower electrodes, respectively, or the lower and upper electrodes, respectively, dependently on the structure of the organic EL element.

① Anode layer

[0078] It is preferred to use, for the anode layer, a metal, an alloy or an electrically conductive compound having a large work function (for example, 4.0 eV or more), or a mixture thereof. Specifically, it is preferred to use one or a combination of two or more selected from indium tin oxide (ITO), indium zinc oxide (IZO), indium copper (CuIn), tin oxide (SnO_2), zinc oxide (ZnO), gold, platinum, palladium and so on.
[0079] By using these electrode materials, the anode layer having a uniform thickness can be made by a method making film-deposition in a dry state possible, such as vacuum deposition, sputtering, ion plating, electron beam evaporation, CVD (chemical vapor deposition), MOCVD (metal oxide chemical vapor deposition), or plasma CVD.
[0080] In the case that EL emission is taken out from the side of the anode layer, it is necessary to make the anode layer to a transparent electrode. On the other hand, in the case that EL emission is not taken out, it is unnecessary to make the anode electrode to a transparent electrode. Therefore, in the case that the anode layer is made to a transparent electrode, it is preferred to use a transparent conductive material such as ITO, IZO, CuIn, SnO_2, or ZnO, and provide the transmittance of EL emission to a value of 70% or more.
[0081] The film thickness of the anode layer is not particularly limited. The thickness is preferably in the range of, for example, 10 to 1,000 nm, and is more preferably in the range of 10 nm to 200 nm.
[0082] The reason for this is as follows: by setting the film thickness of the anode layer in such a range, good electrically connecting reliability can be obtained between the anode layer and electrically connecting members made of IZO, and

further such a film thickness makes it possible to obtain the transmittance of EL emission, for example, 70% or more.

[0083] In the case that EL emission is taken out from the side of the anode layer, it is preferred to set the refractive index of the anode layer in the range of 1.6 to 2.2. This is because, by setting the refractive index in such a range, it is possible to satisfy the above-mentioned expressions (1) and so on easily, and anode materials which can be used are selected from wider range of materials.

[0084] It is therefore more preferred to set the refractive index of the anode layer in the range of 1.7 to 2.1.

[0085] In order to make the adjustment of the relationships about the refractive indexes easy, it is more preferred to use an indium zinc oxide (refractive index: 2.1) among the above-mentioned constituent materials of the anode layer.

② Cathode layer

[0086] It is preferred to use, for the cathode layer, a metal, an alloy or an electrically conductive compound having a small work function (for example, less than 4.0 eV), a mixture thereof, or an inclusion thereof.

[0087] Specifically, it is preferred to use one or a combination of two or more selected from sodium, sodium-potassium alloy, cesium, magnesium, lithium, magnesium-silver alloy, aluminum, aluminum oxide, aluminum-lithium alloy, indium, rare earth metals, mixtures of any one of these metals and an organic luminescence medium material, mixtures of any one of these metals and an electron injection layer material, and so on.

[0088] The film thickness of the cathode layer is not particularly limited in the same way as in the anode layer. Specifically, the thickness is preferably in the range of 10 to 1,000 nm, and is more preferably in the range of 10 nm to 200 nm.

[0089] The reason for this is as follows: by setting the film thickness of the cathode layer in such a range, good electrically connecting reliability can be obtained between the cathode layer and electrically connecting members made of IZO, and further such a film thickness makes it possible to obtain the transmittance of EL emission, for example, 10% or more, more preferably 70% or more.

[0090] In the case that light is taken out from the side of the cathode layer, it is preferred to set the refractive index of the cathode layer in the range of 1.6 to 2.2 in the same way as in the case of the anode layer.

(3) Inter-insulator

[0091] The interlayer dielectric in the organic EL display device of the present invention is present near or around the organic EL elements and the TFTs, and causes the unevenness of a luminescent medium or a color filter to be flattened, so as to be used mainly as a flattened undercoat when the lower electrode of the organic EL element is formed. The interlayer dielectric is also used to attain electric insulation for forming high resolution wiring materials, electric insulation (prevention of short circuits) between the lower and upper electrodes of the organic EL element, electrical insulation or mechanical protection of the TFT, electrical insulation between the TFT and the organic EL element, and so on.

[0092] Therefore, the inter-insulator may be called a flattening film, an electrically insulating film, a partition, a spacer, or the like if necessary. The present invention embraces all of them.

① Constituent material

[0093] Examples of the constituent material used for the inter-insulator usually include acrylic resin, polycarbonate resin, polyimide resin, fluorinated polyimide resin, benzoguanamine resin, melamine resin, cyclic polyolefin, Novolak resin, polyvinyl cinnamate, cyclic rubber, polyvinyl chloride resin, polystyrene, phenol resin, alkyd resin, epoxy resin, polyurethane resin, polyester resin, maleic acid resin, and polyamide resin.

[0094] In the case that the inter-insulator is composed of an inorganic oxide, examples of preferred oxides include silicon oxide (SiO_2 or SiO_x), aluminum oxide (Al_2O_3 or AlO_x), titanium oxide (TiO_2), yttrium oxide (Y_2O_3 or YO_x), germanium oxide (GeO_2 or GeO_x), zinc oxide (ZnO), magnesium oxide (MgO or MgO_x), calcium oxide (CaO), boric acid (B_2O_3), strontium oxide (SrO), barium oxide (BaO), lead oxide (PbO), zirconia (ZrO), sodium oxide (Na_2O), lithium oxide (Li_2O), and potassium oxide (K_2O). The symbol x in the structural formulae representing the inorganic oxides is in the range of 1 to 3.

[0095] In the case that heat-resistance is particularly required, it is preferred to use acrylic resin, polyimide resin, fluorinated polyimide, cyclic polyolefin, epoxy resin, or any inorganic oxide among these constituent materials of the inter-insulator.

[0096] These inter-insulator can be worked into a desired pattern by introducing a photosensitive group thereto and using photolithography, or can be formed into a desired pattern by printing.

② Thickness of the inter-insulator, and so on

[0097] The thickness of the inter-insulator depends on the resolution of display, and the degree of the unevenness of

a fluorescent medium or a color filter combined with the organic EL element, and is preferably in the range of 10 nm to 1 mm.
[0098] This is because such a structure makes it possible to make the unevenness of the fluorescent medium or the color filter sufficiently flat or reduce the dependency of the high resolution display on viewing angle.
[0099] Accordingly, the thickness of the inter-insulator is more preferably in the range of 100 nm to 100 μm, and is still more preferably in the range of 100 nm to 10 μm.

4. Sealing member

① Structure of the sealing member

[0100] In order to prevent moisture from invading the inside of the organic luminescent medium 24, it is preferred that the sealing member 58 illustrated in FIG. 2 is covered at least the luminescent area of the organic EL display device 62.
[0101] As such a sealing member, the same material for the supporting substrate can be used. Particularly, a glass plate having a high effect of shielding moisture or oxygen can be used. The form of the sealing member is not particularly limited, and is preferably, for example, a plate form or a cap form. For example, in the case of a plate form, the thickness thereof is preferably in the range of 0.01 to 5 mm.
[0102] It is also preferred that the sealing member is fitted into a groove or the like made in a part of the supporting substrate under pressure and is then fixed thereto, or that the sealing member is fixed to a part of the supporting member with a photo-setting adhesive agent or the like.

② Sealing medium

[0103] The sealing medium is arranged between the sealing member and the organic EL display device. Examples of such a sealing medium include a transparent resin, and a sealing liquid. More specifically, the sealing medium is selected from a transparent resin or sealing liquid in which a fluorene skeleton containing compound, a bromine containing compound, a sulfur containing compound, or alkoxy titanium is added, a transparent resin or sealing liquid in which SiO_xN_y, Si_3N_4 or AlO_xN_y particles are dispersed; or a transparent resin selected from polyphenyl methacrylate, polyethylene terephthalate, poly-o-chlorostyrene, poly-o-naphthyl methacrylate, polyvinyl naphthalene, polyvinyl carbazole, and polyester containing fluorene skeleton.
[0104] The refractive index of the sealing medium is preferably 1.5 or more. The reason is as follows: since the sealing medium contacts the transparent electrode (refractive index: about 1.6 to 2.1), the refractive index of the sealing medium can be made near to that of the transparent electrode by setting the refractive index of the sealing medium 1.5 or more. In this way, reflection of light on the interface therebetween can be suppressed.
[0105] Since reflection of light on the interface between the transparent electrode and the sealing medium can be more suppressed, the refractive index of the sealing medium is more preferably 1.56 or more, and is still more preferably in the range of 1.58 to 2.0.
[0106] It is also preferred to use, as a main component, a fluorene skeleton containing compound, a bromine containing compound, or a sulfur containing compound in the transparent resin or the sealing liquid constituting the sealing medium, or add the same, as a refractive index adjusting agent, thereto. This is because such a compound has a relatively high refractive index and makes it possible to adjust the refractive index flexibly if necessary.
[0107] Furthermore, in the case that the sealing medium is a transparent resin, the resin is preferably a ultraviolet ray setting resin, a visible ray setting resin, a thermosetting resin, or an adhesive agent using such a resin. Specific examples thereof include Luxtrak LCR0278 and 0242D (both of which are made by Toagosei Co., Ltd., TB3102 (epoxy type, made by Three Bond Co., Ltd.) and Venefix VL (acrylic type, made by Adel Co., Ltd.), which are commercially available.
[0108] In this case the transparent resin constituting the sealing medium is as follows:

polyphenyl methacrylate (refractive index: 1.57)
polyethylene terephthalate (refractive index: 1.58)
poly-o-chlorostyrene (refractive index: 1.61)
poly-o-naphthyl methacrylate (refractive index: 1.61)
polyvinyl naphthalene (refractive index: 1.68)
polyvinyl carbazole (refractive index: 1.68)
polyester containing fluorene skeleton (refractive index: 1.61 to 1.64)

[0109] It is also preferred to add alkoxy titanium such as dimethoxy titanium or diethoxy titanium in the transparent resin or the sealing liquid constituting the sealing medium.
[0110] The addition of the alkoxy titanium in this way makes it possible to make the refractive index of the transparent resin or the sealing liquid to a higher value.

[0111] A transparent inorganic material is preferably contained as the sealing medium. Examples of the transparent inorganic material include SiO_xN_y, Si_3N_4, and AlO_xN_y.

[0112] In the case of forming the transparent inorganic material film, the film is preferably formed at a low temperature (100°C or lower) and a slow film-forming speed in order that the organic EL element is not deteriorated. Specifically, a method such as sputtering, vapor deposition or CVD is preferred.

[0113] These transparent inorganic material films are preferably in an amorphous form since the amorphous films have a high effect of shielding moisture, oxygen, a low molecular monomer and so on and suppress the deterioration of the organic EL element.

[0114] In the case of forming a layer wherein the transparent inorganic material is dispersed, a solution wherein transparent inorganic material particles are dispersed in the transparent resin or the sealing liquid is prepared and this solution may be made into a film by spin coating, roll coating or casting. Alternatively, the inorganic material in a liquid form may be injected under the sealing member.

[0115] In the above-mentioned sealing medium, layers made of materials of different kinds may be composed as plural layers.

[0116] In the case that the sealing medium is composed of plural layers but the refractive indexes of the respective layers are indefinite, the average refractive index of the plural layers may be defined as the refractive index of the sealing medium. In this case, however, it is necessary that the magnitude order of the upper electrode (n1), the sealing medium (n2) and the sealing member (n3) is as follows: $n1 \geqq n2 \geqq n3$ 5. Thin film transistor (TFT)

(1) Structure

[0117] One embodiment of the organic EL display device of the present invention has the plural TFTs 14 and the plural organic EL elements 26 driven correspondingly to the TFTs 14 on the substrate 10, as illustrated in FIG. 2.

[0118] As illustrated in FIG. 2, the flattened inter-insulator 13 is arranged between the TFTs 14 and the lower electrodes 22 of the organic EL elements 26, and further the drain 47 of the TFT 14 and the lower electrode 22 of the organic EL element 26 are electrically connected to each other through the contact hole 48 made in the inter-insulator 13.

[0119] A circuit diagram in one example of such an organic EL display device is illustrated in FIG. 3. This circuit diagram illustrates the following: gate lines (scanning electrode lines) and source lines (signal electrode lines) are formed on the substrate so as to be in an XY matrix form. About each pixel, the following are connected to the gate lines and the source lines: two TFTs 55 and 56 and a condenser 57 for holding the gate of the second TFT 55 at a constant voltage. In this manner, the organic EL element 62 can be driven by the second TFT 56.

[0120] FIG. 4 is a perspective view in a plane direction of the organic EL display device according to the circuit diagram illustrated in FIG. 3.

[0121] Preferably, plural scanning electrode lines (Y_j to Y_{j+n}) 50 and signal electrode lines (X_i to X_{i+n}) 51 arranged in an XY matrix form are also electrically connected to the TFTs 55 and 56 so as to make electric switches for driving the organic EL elements 26, as illustrated in FIG. 3.

[0122] That is, the electric switches are composed of the scanning electrode lines and the signal electrode lines which are electrically connected and, for example, the following: the first transistor(s) (which may be referred to as Tr1 hereinafter) 55, the second transistor(s) (which may be referred to as Tr2 hereinafter) and condenser(s) 57, the number of each of which is one or more.

[0123] The first transistor 55 has a function of selecting one out of luminescent pixels, and the second transistor 56 has a function of driving one of the organic EL elements.

[0124] An active layer 44 of each of the first transistor (Tr1) 55 and the second transistor (Tr2) 56 can be represented by $n^+/i/n^+$. As illustrated in FIG. 2, the regions n^+ at the both side are composed of semiconductor regions 45 and 47 doped in an n type, and the region i therebetween is composed of a non-doped semiconductor region 46.

[0125] The semiconductor regions doped in an n type become a source 45 and a drain 47, which constitute each of the transistors 55 and 56 as illustrated in FIG. 4, together with a gate disposed, across a gate oxide film, on the non-doped semiconductor region.

[0126] In the active layer 44, the semiconductor regions 45 and 47, which are doped in an n type, may be doped in a p type instead of the n type so as to produce a structure of $p^+/i/p^+$.

[0127] The active layers 44 of the first transistor (Tr1) 55 and the second transistor (Tr2) 56 are preferably made of an inorganic semiconductor such as polysilicone, or an organic semiconductor such as thiophene oligomer or poly(p-phenylenevinylene). Polysilicon is a particularly preferable material since it has more sufficient stability against electric conduction than amorphous Si (α-Si).

(2) Driving method

[0128] The following will describe the method of driving the organic EL element by the TFT.

[0129] As illustrated in FIG. 3, the TFT comprises the first transistor (Tr1) 55 and the second transistor (Tr2) 56, and further constitutes the electric switch.

[0130] Therefore, by inputting a scanning signal pulse and a signal pulse through an X-Y matrix to this electric switch to perform switching action, the organic EL element 26 connected to this electric switch can be driven. As a result, the organic EL element 26 gives luminescence or the luminescence is stopped so that image display can be performed.

[0131] That is, by a scanning pulse transmitted through the scanning electrode lines (which may be referred to as gate lines) (Y_j to Y_{j+n}) 50 and a signal pulse transmitted through the signal electrode lines (X_i to X_{i+n}) 51, a desired one out of the first transistors (Tr1) 55 is selected so that a given amount of charges is given into the condenser 57 formed between common electrode lines (C_i to C_{i+n}) 52 and the first transistor (Tr1) 55, as illustrated in FIG. 3.

[0132] In this way, the gate voltage of the corresponding second transistor (Tr2) 56 becomes a constant value so that the second transistor (Tr2) 56 turns on. In this ON-state, the gate voltage is held until the next gate pulse is transmitted. Therefore, electric current is continuously supplied to the lower electrode 22 of the organic EL element 26 connected to the drain of the second transistor (Tr2) 56.

[0133] The organic EL element 26 is driven by the supplied electric current. Thus, the driving voltage thereof is largely reduced and the luminescent efficiency thereof is improved. Moreover, the power consumption thereof can be reduced.

6. Electrically connecting member

[0134] In the first embodiment, the electrically connecting member is preferably made of a noncrystalline conductive oxide, for example, indium zinc oxide (IZO), as well as a metal material.

[0135] That is, due to the characteristics of the noncrystalline conductive oxide, such as moisture resistance and heat resistance, good electric connection can be obtained between the organic EL element and the TFT.

[0136] And due to the excellent etching property that the noncrystalline conductive oxide has, the electrically connecting member superior in precision can easily be formed.

[0137] Furthermore, the noncrystalline conductive oxide has a characteristic of being superior in the capability of electrical connection to the transparent electrode.

[0138] The noncrystalline conductive oxide preferably contains, as a dopant or dopants for adjusting electric conductivity, for example, one or a combination of two or more selected from Sn, Sb, Ga, Ge and so on.

(2) Color filter

① Structure

[0139] The color filter is set in order to decompose or cut light rays to adjust color or improve contrast, and is comprising of a colorant layer consisting only of a colorant or a layer wherein a colorant is dissolved or dispersed in a binder resin.

[0140] The structure of the color filter preferably comprises blue, green and red colorants. This is because combination of such a color filter with the organic EL element giving white luminescence makes it possible to obtain the three primary colors of light, blue, green and red colors, and attain full color display.

[0141] The color filter is preferably patterned by printing or photolithography as in the fluorescent medium.

② Thickness of the color filter

[0142] The thickness of the color filter is not particularly limited if the thickness causes luminescence of the organic EL element to be sufficiently received (absorbed) and does not block color changing function. For example, the thickness is preferably in the range of 10 nm to 1,000 μm, is more preferably in the range of 0.5 pm to 500 μm, and is still more preferably in the range of 1 μm to 100 μm.

(3) Fluorescent medium

① Structure

[0143] The fluorescent medium in the organic EL display device has a function of absorbing luminescence of the organic EL element to give fluorescence having longer wavelengths, and is comprising of layer arranged separately in plane. Each of the fluorescent medium layers is preferably arranged correspondingly to the luminescent area of the organic EL element, for example, the position of the portion where the lower electrode and the upper electrode cross each other. In the case that the organic luminescent layer at the portion where the lower electrode and the upper electrode cross each other gives light, such a structure as above makes it possible that the respective fluorescent medium layers receive the light so that luminescence rays having different colors (wavelengths) can be taken out. A structure wherein the organic EL element emits blue light and further the light can be changed to green and red luminescence rays by the fluorescent medium is particularly preferred since the three primary colors of light, blue, green and red colors, can be obtained even from the single organic EL element so that full color display can be attained.

② Constituent material

[0144] The constituent material of the fluorescent medium is not particularly limited and is made of, for example, a fluorescent dye and a resin, or only a fluorescent dye. The fluorescent dye and the resin may be those in a solid state wherein the fluorescent dye is dissolved or dispersed in a pigment resin and/or a binder resin.

[0145] Specific examples of the fluorescent dye will be described. Examples of the fluorescent dye for changing rays within the range from near-ultraviolet rays to violet luminescence rays in the organic EL element to blue luminescence rays include stylbene colorants such as 1,4-bis(2-methylstyryl)benzene (referred to as Bis-MBS hereinafter) and trans-4,4'-diphenylstylbene (referred to as DPS hereinafter); and coumarin dyes such as 7-hydroxy-4-methylcoumarin (referred to as coumarin 4 hereinafter).

[0146] Examples of the fluorescent dye for changing blue, bluish green or white luminescence rays in the organic EL element to green luminescence rays include coumarin dye such as 2,3,5,6-1H,4H-tetrahydro-8-trifluoromethylquinolidino (9,9a,1-gh)coumarin (referred to as coumarin 153 hereinafter), 3-(2'-benzothiazolyl)-7-diethylaminocoumarin (referred to as coumarin 6) and 3-(2'-benzimidazolyl)-7-N,N-diethylaminocoumarin (referred to as coumarin 7 hereinafter); Basic Yellow 51, which is a coumarin colorant type dye; and naphthalimide dyes such as Solvent Yellow 11 and Solvent Yellow 116.

[0147] Examples of the fluorescent colorant for changing luminescence rays within blue to green luminescence rays, or white luminescence rays in the organic EL element to luminescence rays within orange to red luminescence rays include cyanine dyes such as 4-dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (referred to as DCM hereinafter); pyridine dyes such as 1-ethyl-2-(4-(p-dimethylaminophenyl)-1,3-butadienyl)-pyridinium-perchlorate (referred to as pyridine 1); rhodamine dyes such as Rhodamine B and Rhodamine 6G; and oxadine colorants.

[0148] Various dyes (direct dyes, acidic dyes, basic dyes, disperse dyes and so on) can be selected as fluorescent colorants if they have fluorescent property.

[0149] The fluorescent dye may be beforehand kneaded into the following pigment resin to be made into a pigment: polymethacrylic acid ester, polyvinyl chloride, vinyl chloride vinyl acetate copolymer, alkyd resin, aromatic sulfonamide resin, urea resin, melamine resin, benzoguanamine resin or the like.

[0150] The binder resin is preferably a material having transparency (light transmittance to visible rays: 50% or more). Examples thereof are transparent resins (polymers) such as polymethyl methacrylate, polyacrylate, polycarbonate, polyvinyl alcohol, polyvinyl pyrrolidone, hydroxyethylcellulose, and carboxymethylcellulose.

[0151] In order to arrange the fluorescent medium separately in plane, a photosensitive resin to which photolithography can be applied may be selected. Examples thereof are photo-setting resist materials having reactive vinyl groups, such as acrylic acid type, methacrylic acid type, polyvinyl cinnamate type and cyclic rubber type materials. In the case that printing is used, a printing ink (medium) using a transparent resin is selected. For example, the following may be used: a monomer, an oligomer or a polymer of polyvinyl chloride resin, melamine resin, phenol resin, alkyd resin, epoxy resin, polyurethane resin, polyester resin, maleic acid resin or polyamide resin; or a transparent resin such as polymethyl methacrylate, polyacrylate, polycarbonate, polyvinyl alcohol, polyvinyl pyrrolidone, hydroxyethylcellulose or carboxymethylcellulose.

③ Refractive index

[0152] For reference, the refractive indexes of materials constituting preferred fluorescent medium are as follows:

vinyl chloride resin : 1.54
vinylidene chloride resin : 1.60
vinyl acetate resin : 1.45
polyethylene resin : 1.51
polystyrene resin : 1.59
methyl methacrylate resin : 1.49
melamine resin : 1.60

[0153] It is proved that these refractive indexes change by dissolving or dispersing a dye for the fluorescent medium (or a colorant for the color filter). In the present invention, therefore, the refractive index can be adjusted by selecting a suitable material appropriately.

④ Forming method

[0154] In the case that the fluorescent medium is made mainly of a fluorescent dye, a film thereof is preferably formed by vacuum deposition or sputtering through a mask for obtaining a desired fluorescent medium pattern.

[0155] On the other hand, in the case that the fluorescent medium is made of a fluorescent dye and a resin, the fluorescent medium is preferably formed by mixing the fluorescent dye and the resin with an appropriate solvent or dispersing or dissolving the fluorescent dye and the resin in the solvent to prepare a liquid material, making the liquid material into a film by spin coating, roll coating, casting or the like and subsequently patterning the film into a desired pattern by photolithography or screen printing or the like.

⑤ Thickness

[0156] The thickness of the fluorescent medium is not particularly limited if the thickness causes luminescence of the organic EL element to be sufficiently received (absorbed) and does not block fluorescence generating function. For example, the thickness is preferably in the range of 10 nm to 1,000 μm, is more preferably in the range of 0.1 μm to 500 μm, and is still more preferably in the range of 5 μm to 100 μm.

[0157] The reason for this is as follows. If the thickness of the fluorescent medium is less than 10 nm, the mechanical strength thereof may drop or the lamination thereof may become difficult. On the other hand, if the thickness of the fluorescent medium is over 1 mm, the light transmittance thereof drops remarkably so that the quantity of light which can be taken out may drop or the organic EL display device may not be easily made thin.

Examples

[Example 1]

(1) Manufacture of fluorescent medium

[0158] A ball mill was used to mix homogeneously 100 g of an acrylic type photo-setting resist 259PA having a fluorene skeleton (made by Shin Nippon Steel chemical Co., Ltd. Solid content: 50% by weight, and propylene glycol ether methyl ether acetate was used as a solvent) as a photo-setting resin, 0.53 g of coumarin 6 as an organic fluorescent dye, 1.5 g of Basic Violet 11, 1.5 g of Rhodamine 6G and 25 g of propylene glycol methyl ether acetate as a solvent, so as to prepare a composition for a fluorescent medium (an ink for a fluorescent medium).

[0159] The resultant fluorescent medium composition was applied onto a glass substrate (Corning 7059) 25 mm in length, 75 mm in width and 1.1 mm in thickness by spin coating, and then dried at 80°C for 10 minutes. Next, ultraviolet rays (wavelength: 365 nm) were radiated thereon in such a manner that the quantity of light exposure would be 1,500 mJ/cm^2, to form the fluorescent medium.

[0160] The refractive index of the fluorescent medium was measured, so that it was 1.62. In the same way, the refractive index of the glass substrate was measured, so that it was 1.50.

[0161] In this way, a sealing member in which the fluorescent medium was formed was manufactured.

(2) Manufacture of organic EL elements

[0162] A glass substrate (Corning 7059) 25 mm in length, 75 mm in width and 1.1 mm in thickness was cleaned with isopropyl alcohol and ultraviolet rays, and subsequently this substrate was fixed onto a substrate holder inside a vacuum deposition machine (ULVAC, Co., Ltd.).

[0163] Next, a heating board made of molybdenum inside the vacuum evaporation machine was filled with 4,4',4"-tris [N-(3-methylphenyl)-N-phenylamino]triphenylamine (MTDATA) and 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPD) as hole injection materials, 4,4'-bis(2,2-diphenylvinyl)biphenyl (DPVBi) as an organic luminescent material, and tris(8-quinolynol)aluminum (Alq) as an electron injection material, and further an Al/Li alloy (Li content by percentage: 5% by weight) as a constituent material of lower electrodes (cathodes) was fitted to the heating board.

[0164] In this state, the evacuated pressure of the deposition machine was reduced to 655 $\times$ 10^{-7} Pa and then the above-mentioned materials were deposited by single vacuum drawing operation, without canceling the vacuum state on its way from the formation of the cathodes to the formation of a hole injection layer, in such a manner that the following deposition rates and film thicknesses would be attained.

MTDATA: deposition rate: 0.1 to 0.3 nm/sec., film thickness: 60 nm,
NPD: deposition rate: 0.1 to 0.3 nm/sec., film thickness: 20 nm,
DPVBi: deposition rate: 0.1 to 0.3 nm/sec., film thickness: 50 nm,

Alq: deposition rate: 0.1 to 0.3 nm/sec., film thickness: 20 nm, and
Al/Li alloy: deposition rate: 1.0 to 2.0 nm/sec., film thickness: 150 nm.

[0165] Next, the substrate was moved to a sputtering machine, and then IZO (refractive index: 2.1) for upper electrodes was sputtered into a film to have a thickness of 200 nm. In this way, organic EL elements were manufactured.

(3) Sealing step

[0166] The organic EL elements obtained in the above-mentioned (2) were put into a dry box which dry nitrogen was introduced. O-PET resin (refractive index: 1.63), which is a polyester resin having a fluorene skeleton, was laminated on the luminescent faces (on the upper electrodes) of the organic EL elements so as to form a film of the sealing medium.
[0167] Next, the glass substrate on which the fluorescent medium (refractive index: 1.62) obtained in the above-mentioned (1) was formed, that is, the sealing member (refractive index: 1.5) was laminated on the sealing medium.
[0168] In other words, the refractive index (n1: 2.1) of the upper electrodes in the organic EL elements, the refractive index (n2: 1.63) of the sealing medium, the refractive index (n4: 1.62) of the first color changing medium, and the refractive index (n3: 1.5) of the sealing member were arranged in this way to satisfy the expression (2).
[0169] The peripheral portion thereof was treated with a cation-setting adhesive agent TB3102 (made by Three Bond Co., Ltd.), and then the agent was set by light to make the resultant workpiece airtight. In this way, an organic EL display device of Example 1 was manufactured.

(4) Evaluation of the organic EL display device

[0170] In Example 1. for evaluation of the invention, a DC voltage of 12 V was applied through an active matrix circuit between the upper electrodes (anodes, IZO) of the resultant organic EL display device and the lower electrodes (cathode, Al/Li) thereof to give luminescence.
[0171] A chroma meter CS1000 (made by Minolta Co., Ltd.) was used to measure the luminescent brightness. As a result, a value of 62 cd/m^2 was obtained. It was proved that the CIE chromaticity coordinates of the resultant red luminescence were as follows: X = 0.62 and Y = 0.34.

[Comparative Example 1]

[0172] In Comparative Example 1, a comparative example was evaluated. Accordingly, in Comparative Example 1, an organic EL display device was manufactured and evaluated in the same manner as in Example 1 except that a silicone oil (refractive index: 1.55) was filled and used instead of the O-PET resin used in Example 1. That is, an organic EL display device was formed in the manner that the refractive index (n1) of the upper electrodes, that (n2) of the sealing medium, that (n4) of the color changing medium and that (n3) of the sealing member did not satisfy the relational expression (2).
[0173] Next, in the same way as in Example 1, the luminescent brightness of the resultant organic EL display device was measured with chroma meter CS1000. As a result, a value of 55 cd/m^2 was obtained as shown in Table 1. Red EL emission was obtained. It was proved that the CIE chromaticity coordinates thereof were as follows: X = 0.62 and Y = 0.34.
[0174] Thus, in Comparative Example 1 the same organic EL elements as in Example 1 were used, but it was proved that the luminescent brightness was reduced by about 11%.
[0175] It was demonstrated that when such silicone oil was used, the organic EL elements were broken out after several minutes from the measurement of the luminescent brightness so that EL emission could not be obtained.

[Table 1]

	Side from which luminescence was taken out	n1	n2	n4	n3	Luminescent brightness (cd/m^2)
Example 1	Upper electrode side	2.1	1.63	1.62	1.50	62
Comparative Example 1	Upper electrode side	2.1	1.55	1.62	1.50	55
Comparative Example 2	Upper electrode side	2.1	1.30	1.62	1.50	50

(continued)

	Side from which luminescence was taken out	n1	n2	n4	n3	Luminescent brightness (cd/m^2)
Comparative Example 3	Upper electrode side	2.1	1.00	1.62	1.50	29

[Comparative Example 2]

[0176] In Comparative Example 2, a further comparative example was evaluated. Accordingly, in Comparative Example 2, an organic EL display device was manufactured and evaluated in the same manner as in Example 1 except that fluorinated hydrocarbon liquid Florinate (made by Sumitomo 3M Limited, refractive index: 1.3) was filled and used instead of the O-PET resin in Example 1. That is, an organic EL display device was formed in the manner that the respective refractive indexes did not satisfy the expression (2).

[0177] Next, a DC voltage of 12 V was applied through an active matrix circuit between the upper electrodes (anodes, IZO) of the resultant organic EL display device and the lower electrodes (cathodes, Al/Li) thereof. The chroma meter CS1000 was used to measure the luminescent brightness.

[0178] As a result, a luminescent brightness value of 50 cd/m^2 was obtained. It was proved that the CIE chromaticity coordinates of the resultant red luminescence were as follows: X = 0.62 and Y = 0.34.

[0179] Thus, in Comparative Example 2 the same organic EL elements as in Example 1 were used, but it was proved that the luminescent brightness was reduced by about 19%.

[Comparative Example 3]

[0180] In Comparative Example 3, yet a further comparative example was evaluated. Accordingly, in Comparative Example 3, an organic EL display device was manufactured and evaluated in the same manner as in Example 1 except that dry nitrogen (refractive index: 1.0) was filled and used instead of the O-PET resin in Example 1. That is, an organic EL display device was formed in the manner that the respective refractive indexes did not satisfy the relational expression (2).

[0181] Next, a DC voltage of 12 V was applied through an active matrix circuit between the upper electrodes (anodes, IZO) of the resultant organic EL display device and the lower electrodes (cathodes, Al/Li) thereof. The chroma meter CS1000 was used to measure the luminescent brightness.

[0182] As a result, a luminescent brightness value of 29 cd/m^2 was obtained. It was proved that the CIE chromaticity coordinates of the resultant red luminescence were as follows: X = 0.62 and Y = 0.34.

[0183] Thus, in Comparative Example 3 the same organic EL elements as in Example 1 were used, but it was proved that the luminescent brightness was reduced by about 50%.

[Example 2]

[0184] In Example 2, the invention was evaluated. That is, an organic EL display device was manufactured under the same conditions as in Example 1 except that a sealing member having no fluorescent medium was used.

[0185] Namely, an organic EL display device was formed in the manner that the refractive index (n1: 2.1) of the upper electrodes in the organic EL element, that (n2: O-PET resin 1.63) of the sealing medium, and that (n3: 1.5) of the sealing member satisfied the expression (1).

[0186] Next, a DC voltage of 12 V was applied through an active matrix circuit between the upper electrodes (anodes, IZO) of the resultant organic EL display device and the lower electrodes (cathodes, Al/Li) thereof to give luminescence.

[0187] The chroma meter CS1000 (made by Minolta Co., Ltd.) was used to measure the luminescent brightness. As a result, a value of 200 cd/m^2 was obtained. It was proved that the CIE chromaticity coordinates of the resultant blue luminescence were as follows: X = 0.15 and Y = 0.16.

[Comparative Example 4]

[0188] In Comparative Example 4, a comparative example was evaluated. Accordingly, in Comparative Example 4 an organic EL display device was manufactured and evaluated in the same manner as in Example 2 except that nitrogen gas (refractive index: 1.00) was used as a sealing medium instead of the O-PET resin used in Example 2. That is, an organic EL display device was formed in the manner that refractive index (n1) of the upper electrodes, that (n2) of the sealing medium and that (n3) of the sealing member did not satisfy the expression (1).

[0189] Next, the chroma meter CS1000 was used to measure the luminescent brightness of the resultant organic EL display device in the same manner as in Example 2. As a result, a value of 160 cd/m^2 was obtained, as shown in Table 2. Blue luminescence was obtained. It was proved that the CIE chromaticity coordinates thereof were as follows: X = 0.15 and Y = 0.16.

[0190] Thus, in Comparative Example 4 the same organic EL elements as in Example 2 were used, but it was proved that the luminescent brightness was reduced by about 20%.

[0191] It was demonstrated that when nitrogen gas was used as the sealing medium in this way, the luminescent brightness dropped remarkably since the expression (1) was not satisfied.

[Table 2]

	Side from which luminescence was taken out	n1	n2	n3	Luminescent brightness (cd/m^2)
Example 2	Upper electrode side	2.1	1.63	1.50	200
Comparative Example 4	Upper electrode side	2.1	1.00	1.50	160

Industrial Applicability

[0192] According to the invention (organic EL display device), the relationship between the refractive index of the sealing member and those of the electrode and so on is considered. In this manner, reflection on the respective interfaces is suppressed and the quantity of EL emission which can be taken out can be made large even if silicone oil or the like is not used as the sealing medium.

Claims

1. An organic electroluminescence display device comprising;
a supporting substrate;
an organic electroluminescence element comprising an organic luminescent medium sandwiched between a lower electrode and an upper electrode on the supporting substrate;
a sealing medium and
a sealing member;
whereby the sealing medium is arranged between the organic electroluminescence element and the sealing member; and
the electroluminescence emission is taken out from the upper electrode; and
the following expression (1) is satisfied

$$n1 \geq n2 \geq n3 \qquad (1)$$

wherein n 1 represents the refractive index of the upper electrode, n2 represents the refractive index of the sealing medium, and n3 represents the refractive index of the sealing member,
characterized in that the sealing medium is:

- a transparent resin or sealing liquid in which a fluorene skeleton containing compound, a bromine containing compound, a sulfur containing compound, or alkoxy titanium is added,
- a transparent resin or sealing liquid in which SiO_xNy, Si_3N_4 or AlO_xN_y particles are dispersed; or
- a transparent resin selected from polyphenyl methacrylate, polyethylene terephthalate, poly-o-chlorostyrene, poly-o-naphthyl methacrylate, polyvinyl naphthalene, polyvinyl carbazole, and polyester containing fluorene skeleton.

2. The organic electroluminescence display device according to claim 1, wherein the device further comprises a color changing medium between the sealing medium and the sealing member; and
the following expression (2) is satisfied

$$n1 \geq n2 \geq n4 \geq n3 \quad (2)$$

wherein n4 represents the refractive index of the color changing medium.

3. The organic electroluminescence display device according to claim 1, wherein the device further comprises a color changing medium on a surface of the sealing member, the surface opposite to a surface on
which the sealing medium is arranged; and
the following expression (3) is satisfied

$$n1 \geq n2 \geq n3 \geq n4' \quad (3)$$

wherein n4' represents the refractive index of the color changing medium.

4. The organic electroluminescence display device according to any one of claims 1 to 3, wherein the refractive indexes n1 and n2 satisfy the following expression (4)

$$n1 \geq n2 \geq 0.7 \times n1 \quad (4).$$

5. The organic electroluminescence display device according to any one of claims 1 to 4, wherein the refractive index of the sealing medium is 1.56 or more.

6. The organic electroluminescence display device according to any one of claims 1 to 5, wherein the sealing medium is a transparent resin or sealing liquid in which SiO_xN_y, Si_3N_4 or AlO_xN_y particles are dispersed.

7. The organic electroluminescence display device according to any one of claims 1 to 6, wherein the upper electrode is made mainly of an indium zinc oxide.

8. The organic electroluminescence display device according to any one of claims 1 to 7, wherein a thin film transistor for driving the organic electroluminescence element is arranged on the supporting substrate.

Patentansprüche

1. Organische Elektrolumineszenz-Display-Vorrichtung, umfassend:

ein Trägersubstrat;
ein organisches Elektrolumineszenzelement, umfassend ein organisches lumineszierendes Medium, das sandwichartig zwischen einer unteren Elektrode und einer oberen Elektrode auf dem Trägersubstrat angeordnet ist;
ein Versiegelungsmedium und
ein Versiegelungsbauteil;
wobei das Versiegelungsmedium zwischen dem organischen Elektrolumineszenzelement und dem Versiegelungsbauteil angeordnet ist; und
die Elektrolumineszenzemission aus der oberen Elektrode entnommen wird; und
die folgende Beziehung (1) erfüllt ist

$$n1 \geq n2 \geq n3 \quad (1)$$

wobei n1 den Brechungsindex der oberen Elektrode darstellt, n2 den Brechungsindex des Versiegelungsmediums darstellt, und n3 den Brechungsindex des Versiegelungsbauteils darstellt,
dadurch charakterisiert, dass das Versiegelungsmedium:

- ein transparentes Harz oder eine Versiegelungsflüssigkeit, zu dem/der eine Fluorengerüst-haltige Verbindung, eine Brom-haltige Verbindung, eine Schwefel-haltige Verbindung oder Alkoxytitan gegeben wird,

- ein transparentes Harz oder eine Versiegelungsflüssigkeit, in dem/der SiO_xN_y-, Si_3N_4- oder AlO_xN_y-Partikel dispergiert sind; oder
- ein transparentes Harz, ausgewählt aus Polyphenylmethacrylat, Polyethylenterephthalat, Poly-O-chlorstyrol, Poly-O-naphthylmethacrylat, Polyvinylnaphthalin, Polyvinylcarbazol und Fluorengerüsthaltiger Polyester ist.

2. Organische Elektrolumineszenz-Display-Vorrichtung nach Anspruch 1 , wobei die Vorrichtung darüber hinaus ein farbänderndes Medium zwischen dem Versiegelungsmedium und dem Versiegelungsbauteil umfasst; und die folgende Beziehung (2) erfüllt ist

$$n1 \geq n2 \geq n4 \geq n3 \qquad (2)$$

wobei n4 den Brechungsindex des farbändernden Mediums darstellt.

3. Organische Elektrolumineszenz-Display-Vorrichtung nach Anspruch 1, wobei die Vorrichtung darüber ein farbänderndes Medium auf einer Oberfläche des Versiegelungsbauteils umfasst, wobei die Oberfläche entgegengesetzt zur Oberfläche angeordnet ist, auf der das Versiegelungsmedium angeordnet ist, und die folgende Beziehung (3) erfüllt ist

$$n1 \geq n2 \geq n3 \geq n4' \qquad (3)$$

wobei n4’ den Brechungsindex des farbändernden Mediums darstellt.

4. Organische Elektrolumineszenz-Display-Vorrichtung nach einem der Ansprüche 1 bis 3, wobei die Brechungsindizes n1 und n2 die folgende Beziehung (4) erfüllen

$$n1 \geq n2 \geq 0{,}7 \times n1 \qquad (4)$$

5. Organische Elektrolumineszenz-Display-Vorrichtung nach einem der Ansprüche 1 bis 4, wobei der Brechungsindex des Versiegelungsmediums 1,56 oder mehr beträgt.

6. Organische Elektrolumineszenz-Display-Vorrichtung nach einem der Ansprüche 1 bis 5, wobei das Versiegelungsmedium ein transparentes Harz oder eine Versiegelungsflüssigkeit ist, in dem/der SiO_xN_y-, Si_3N_4- oder AlO_xN_y-Partikel dispergiert sind.

7. Organische Elektrolumineszenz-Display-Vorrichtung nach einem der Ansprüche 1 bis 6, wobei die obere Elektrode hauptsächlich aus einem Indiumzinkoxid hergestellt ist.

8. Organische Elektrolumineszenz-Display-Vorrichtung nach einem der Ansprüche 1 bis 7, wobei ein Dünnfilmtransistor zum Antrieb des organischen Elektrolumineszenzelements auf dem Trägersubstrat angeordnet ist.

Revendications

1. Dispositif d’affichage par électroluminescence organique comprenant :

un substrat de support,
un élément électroluminescent organique comprenant un milieu luminescent organique coincé entre une électrode inférieure et une électrode supérieure sur le substrat de support,
un milieu d’étanchéité et
un membre d’étanchéité ;
où le milieu d’étanchéité est arrangé entre l’élément électroluminescent organique et le membre d’étanchéité ; et l’émission électroluminescente est prise de l’électrode supérieure ; et l’expression (1) suivante est satisfaire :

$$n1 \geq n2 \geq n3 \qquad (1)$$

dans laquelle n1 représente l'indice de réfraction de l'électrode supérieure, n2 représente l'indice de réfraction du milieu d'étanchéité et n3 représente l'indice de réfraction du membre d'étanchéité,
caractérisé en ce que le milieu d'étanchéité est :

- une résine transparente ou un liquide d'étanchéité, à qui un composé contenant un squelette de fluorène, un composé contenant du brome, un composé contenant du souffre ou un alcoxy titane est ajouté,
- une résine transparente ou un liquide d'étanchélté, dans laquelle/lequel des particules de SiO_xN_y, de Si_3N_4 ou de AlO_xN_y sont dispersées ; ou
- une résine transparente sélectionnée parmi du méthacrylate de polyphényle, du téréphtalate de polyéthylène, du poly-o-chlorostyrène, du méthacrylate de poly-o-naphtyle, du polyvinyle naphtalène, du polyvinyle carbazole et du polyester contenant un squelette de fluorène.

2. Dispositif d'affichage par électroluminescence organique selon la revendication 1, dans lequel le dispositif comprend en outre un milieu de changement de couleur entre le milieu d'étanchéité et le membre d'étanchéité ; et l'expression (2) suivante est satisfaite :

$$n1 \geq n2 \geq n4 \geq n3 \qquad (2)$$

dans laquelle n4 représente l'indice de réfraction du milieu de changement de couleur,

3. Dispositif d'affichage par électroluminescence organique selon la revendication 1, dans lequel le dispositif comprend en outre un milieu de changement de couleur sur une surface du membre d'étanchéité, la surface opposée d'une surface sur laquelle le milieu d'étanchéité est arrange ; et
l'expression (3) suivante est satisfaite :

$$n1 \geq n2 \geq n3 \geq n4' \qquad (3)$$

dans laquelle n4' représente l'indice de réfraction du milieu de changement de couleur,

4. Dispositif d'affichage par électroluminescence organique selon l'une des revendications 1 à 3, dans lequel les indices de réfraction n1 et n2 satisfont l'expression (4) suivante :

$$n1 \geq n2 \geq 0{,}7 \times n1 \qquad (4).$$

5. Dispositif d'affichage par électroluminescence organique selon l'une des revendications 1 à 4, dans lequel l'indice de réfraction du membre d'étanchéité est égal ou supérieur à 1,56.

6. Dispositif d'affichage par électroluminescence organique selon l'une des revendications 1 à 5, dans lequel le membre d'étanchéité est une résine transparente ou un liquide d'étanchéité, dans laquelle/lequel des particules de SiO_xN_y, de Si_3N_4 ou de AlO_xN_y sont dispersées.

7. Dispositif d'affichage par électroluminescence organique selon l'une des revendications 1 à 6, dans lequel l'électrode supérieure est principalement faite d'un oxyde d'étain-indium.

8. Dispositif d'affichage par électroluminescence organique selon l'une des revendications 1 à 7, dans lequel un transistor à couche mince pour commander l'élément électroluminescent organique est arrangé sur le substrat de support,

FIG. 1

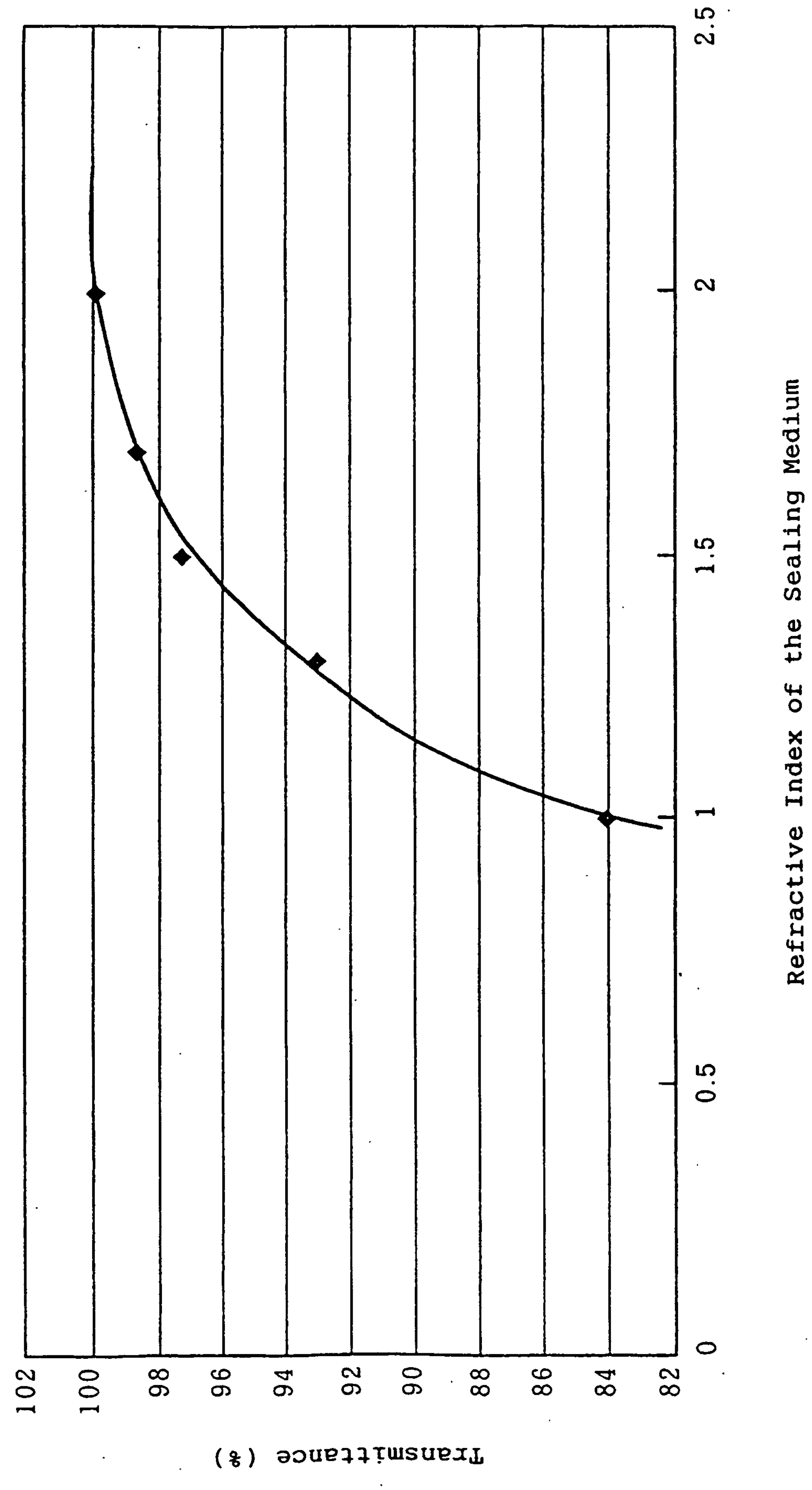

Transmittance (%)
102
100
98
96
94
92
90
88
86
84
82
Refractive Index of the Sealing Medium
0
0.5
1
1.5
2
2.5

FIG. 2

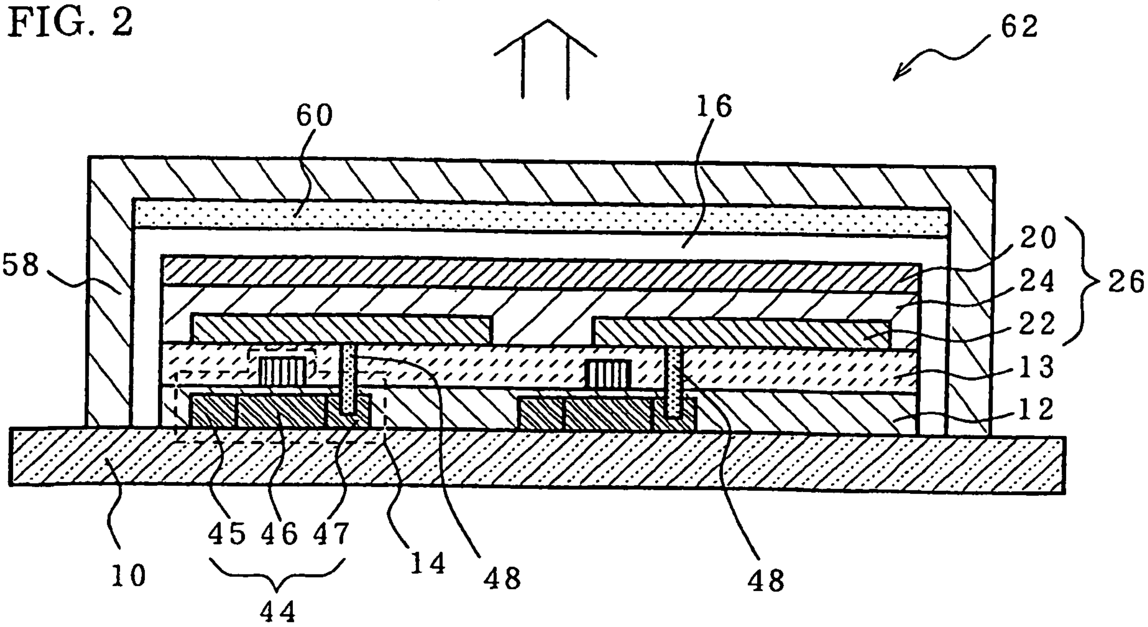
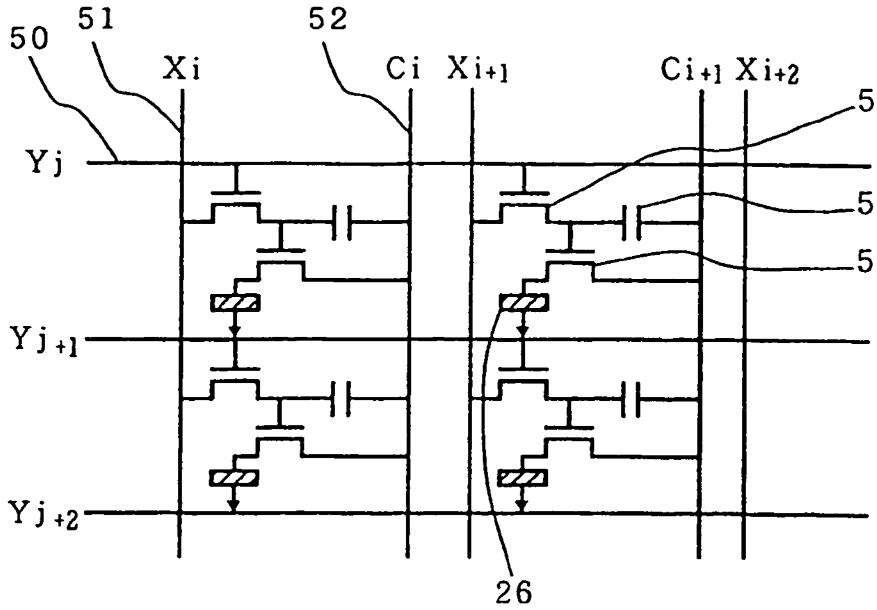


FIG. 3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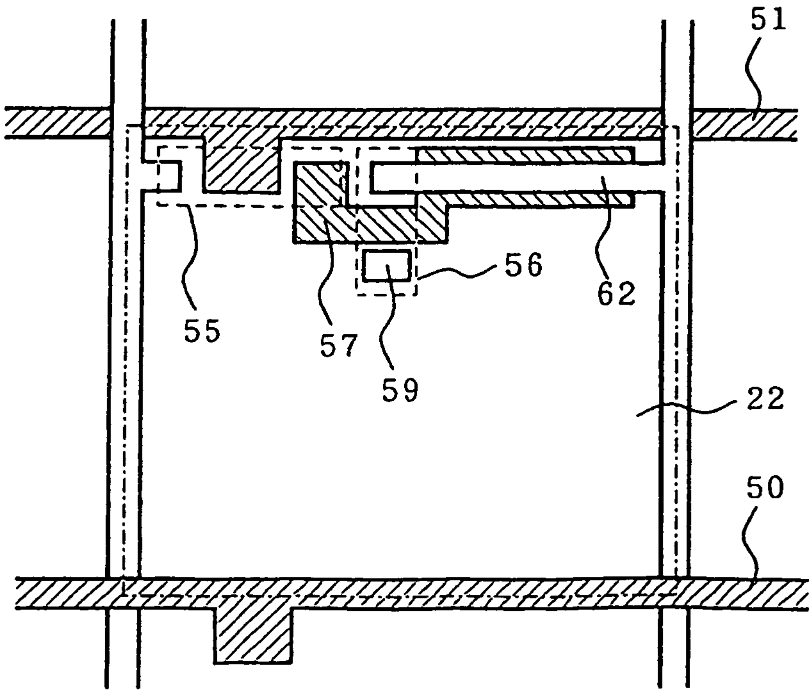
51
Xi
52
Ci
Xi+1
Ci+1
Xi+2
55
57
56
Yj
Yj+1
Yj+2
26

FIG. 4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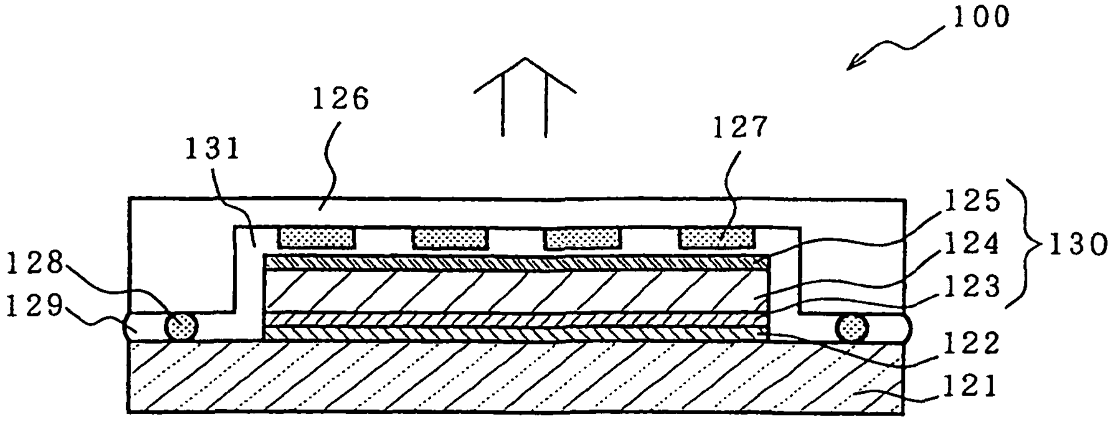
55
57
56
59
62
22
50

FIG. 5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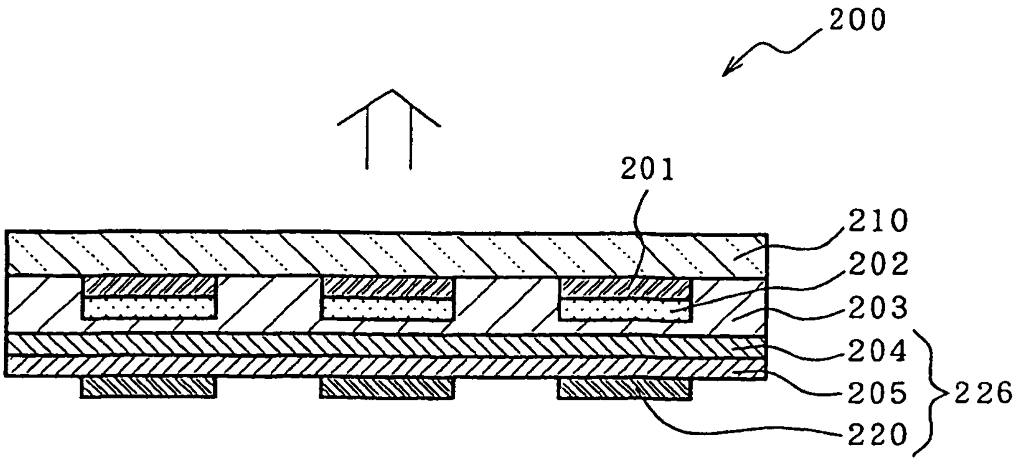
122
121

FIG. 6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220

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- JP HEI10289784 B **[0004] [0011]**
- JP HEI11185955 B **[0004]**
- JP HEI10162958 B **[0006] [0012]**
- JP 11185955 A **[0011]**

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申请(专利权)人(译)	出光兴产股份有限公司.		
当前申请(专利权)人(译)	出光兴产股份有限公司.		
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发明人	EIDA, MITSURU TOMOIKE, KAZUHIRO		
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摘要(译)

1.一种有机EL显示装置，包括：支撑基板;以及包括夹在下电极和上电极之间的有机发光介质的有机EL元件。在支撑基板和下电极之间设置变色介质和/或透明树脂层。从下电极取出EL发射。有机EL显示装置的各种元件的折射率之间表示各种关系。在这种布置中，大量的EL发射可以被带到外部。

$$R = (n_a - n_b)^2/(n_a + n_b)^2$$